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DECREASING FLUORESCENCE OF
SYNTHETIC CAFFEINEJay S. Buckley, Jr., Groton, Conn., assignor to
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This invention relates to the preparation of caffeine of improved quality and particularly to non-fluorescent synthetic caffeine.

Caffeine prepared synthetically often has a considerable bluish fluorescence which appears both in the solid compound and in solutions thereof. Natural caffeine possesses a similar fluorescence but to a much more limited degree. The increased fluorescence of the synthetic material is most undesirable, since it is often transmitted to other products in which the caffeine is utilized, to their marked disadvantage.

Various methods known to be useful in removing impurities from other organic compounds have been tried in an attempt to remove the fluorescing contaminants from synthetic caffeine. Aqueous solutions of the material have been treated with activated carbon, but only slight improvement is obtained. The same is true of treatments with alumina and in alumina columns. Some improvement is realized when a chloroform solution of the caffeine is employed instead of an aqueous solution. However, using a large volume of organic solvents is both expensive and inconvenient, and additional operations of extracting the caffeine from the water solution in which it is normally prepared into the solvent and of later recovering the solvent are required. In addition, the usual precautions must be taken which are necessary when working with such organic materials.

It has now been found that when aqueous or organic solvent solutions of crude synthetic caffeine are treated with nitrous acid either per se or through a precursor of nitrous acid and the caffeine then recovered, fluorescence of the product is effectively and most unexpectedly minimized or obviated. Only a small amount of the reagent is required to decrease the fluorescence of synthetic caffeine to a low value comparable at least to that of natural caffeine. Rather than using nitrous acid as the agent we may use suitable precursors of it such as the oxides of nitrogen which are generated by the action of mineral acids (e. g. hydrochloric acid) on metallic nitrites. A further alternative is the use of organic nitrites such as ethyl, butyl, or amyl nitrite. The lower members of this series are gases and may be generated and piped into the reaction mixture. Certain organic liquids, such as chloroform, may be used as the solvent for the caffeine rather than water, and in this case the use of organic nitrites with an organic acid is indicated. However, the use of aqueous solutions is preferred in large scale operation.

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In the preferred practice of this invention, a warm or hot aqueous solution of the crude synthetic caffeine, at a pH which is slightly acid or neutral, is treated with a weight of a soluble nitrous acid salt, say an alkali metal nitrite, equal to about 1 to 5 per cent of the weight of caffeine present. Either before, during, or after the nitrite treatment, acid is added to the solution in an amount necessary to free the nitrous acid. For instance, acetic acid or a mineral acid, etc., may be used for this purpose. After stirring for a short time, the mixture is neutralized or made slightly basic and then cooled, directly or after some concentration, to crystallize the purified caffeine. It is best to employ solutions of the caffeine ranging in concentration from about 10 to 25 per cent by weight, and it is generally most economical and convenient to use a concentration of about 20 to 25 per cent. For efficient operation the temperature of the aqueous solution should be such that the caffeine is readily soluble; this may vary from 70° to 100° C., depending upon the concentration used, and the preferred range is 80° to 85° C. Before the solution is cooled to crystallize the caffeine, it may be treated with decolorizing carbon to remove colored impurities; however, this has little effect on the fluorescence of the product. The mother liquor left after removal of a crop of caffeine crystals may be concentrated to recover further material, or it may be extracted with a solvent such as chloroform.

According to one specific embodiment of this invention, synthetically prepared caffeine having an undesirably high fluorescence value is dissolved in hot water at a concentration of 200 to 240 grams (as anhydrous caffeine) per liter. The mixture is heated to 80° to 95° C. and adjusted to pH 6-7. This solution is treated with two to four pounds of commercial sodium or potassium nitrite per one hundred pounds of caffeine in the batch. An aqueous solution of acetic acid stoichiometrically equivalent to the nitrite content is then slowly added to the well-stirred mixture over a period of about fifteen minutes or more, while an elevated temperature is maintained. The pH is then adjusted to 7.8 with a caustic soda solution and the hot material treated with activated carbon and a filteraid. After filtration the clear solution is cooled, and crystallized caffeine of markedly lower fluorescence is obtained.

The recovery of purified caffeine from this treatment is excellent, over 95 per cent yield normally being obtained. The quality of the material is greatly improved. Fluorescence of the various samples of caffeine is measured on solutions